

Absence of bimodal peak spacing distribution in the Coulomb blockade regime

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Abstract

Using exact diagonalization numerical methods, as well as analytical arguments, we show that for the typical electron densities in chaotic and disordered dots the peak spacing distribution is not bimodal, but rather Gaussian. This is in agreement with the experimental observations. We attribute this behavior to the tendency of an even number of electrons to gain on-site interaction energy by removing the spin degeneracy. Thus, the dot is predicted to show a non trivial electron number dependent spin polarization. Experimental test of this hypothesis based on the spin polarization measurements are proposed.

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The distribution of the addition spectrum of chaotic and disordered quantum dots measured in recent experiments [1–3] seem to contradict the predictions of the orthodox constant interaction model [4–7]. The most striking feature in the addition spectrum distribution is the absence of any signs of a bimodal structure corresponding to the two spin states of the tunneling electron. In the constant interaction (CI) approximation, the ground state energy of a quantum dot populated by N electrons is given by $E_N = e^2 N^2 / 2C + \sum_{i=1}^{N_\uparrow} \eta_i + \sum_{i=1}^{N_\downarrow} \eta_i$ where C is the dot's constant (or slowly varying) capacitance, η_i are the single particle energies, and N_\uparrow (N_\downarrow) is the number of electrons for which the spin is in the up (down) state ($N = N_\uparrow + N_\downarrow$). In the absence of a magnetic field, for an even number of electrons $N_\uparrow = N_\downarrow = N/2$ while for an odd N , $N_\uparrow = (N+1)/2$ and $N_\downarrow = (N-1)/2$. Thus, the change in the chemical potential needed to add a particle, $\Delta_2 = \mu_N - \mu_{N-1} = E_N - 2E_{N-1} + E_{N-2}$, is equal to e^2/C for N even, while $\Delta_2 = e^2/C + \eta_{(N+1)/2} - \eta_{(N-1)/2}$ for N odd. Assuming that the single electron level spacings follow the random matrix theory (RMT) predictions, the distribution of the spacings should be, in the absence of a magnetic field,

$$P_{CI+RMT}(s) = \frac{1}{2} \left(\delta(s) + \pi s \exp(-\pi s^2) \right), \quad (1)$$

where $s = (\Delta_2 - e^2/C)/2\Delta$, $\Delta = \langle \eta_i - \eta_{i-1} \rangle$ is the averaged single level spacing and $\langle \dots \rangle$ denotes an average over realizations or levels. As we have mentioned, none of the experimental distributions shows even the slightest sign of any bimodal distribution.

When the interactions are treated beyond the CI model, different distributions emerge. In Ref. [8], the influence of the interactions on the distribution within the Hartree approximation was considered. For strong interactions, this, roughly speaking, leads to a distribution composed of two uncorrelated Wigner distributions resulting in the spin resolved (SR) RMT distribution (see [3,9]) which is plotted in Fig. 2a. This distribution begins abruptly ($P(s < 0) = 0$ while $P(s = 0) = 1/2$) and is strongly asymmetric. All the experimental distributions are nevertheless more or less Gaussian with only weak asymmetries in the tails [1–3].

Thus, none of the above models can satisfactorily reproduce the experimental distribu-

tions. We therefore use an exact diagonalization study to investigate the role of spin in determining the addition spectrum of a disordered tight-binding Hamiltonian with on-site and long range interactions. Although we pay the price of handling only small systems, we avoid the uncertainty of approximations and fully include the effects of electron correlations. In similar studies of symmetric dots, atoms and nuclei [6,7,10] the spatial symmetry of the confining potential plays an important role and leads to a strong dependence on the particle numbers via the shell structure (magic numbers) and a non-trivial total spin due to Hund rules. Because of the chaotic nature of the dots [1–3] we do not expect such dependence on the electron number to play an important role, and therefore the study of a small number of electrons is still useful in understanding the properties of dots which are populated by an order of magnitude more electrons. Both components of the Coulomb interaction on a lattice, i.e., the long range part and the on-site interaction must be considered since without the long range component of the Coulomb interaction the classical capacitance behavior for the average spacing will not be reproduced, and for electrons with spin on a lattice a Coulomb interaction must have an on-site component.

Three different regions of behavior emerge. The weak interaction limit, which corresponds to an on-site interaction parameter smaller than a fifth of the bandwidth, is characterized by a strong even-odd asymmetry of the addition spectrum. As discussed below this region corresponds to values of $r_s < 0.1 - 0.3$ (where r_s is the ratio of the inter-particle Coulomb energy to the kinetic energy). An intermediate region corresponding to $0.1 - 0.3 < r_s < 1$, where there is no sign of the spin in the Δ_2 distribution, the distribution tends towards Gaussian, and the total spin of an even number of electrons in the dot is likely to be partially polarized, while the spin of an odd number of electrons is hardly affected and is equal to $\hbar/2$. For the strong interaction regime $r_s > 1$ the distribution is a Gaussian of a width proportional to e^2/C , and the spin polarization of the dot shows a complicated behavior.

Since all experiments are in the $r_s \sim 1$ regime [1,3] these results clearly are consistent with no bimodal distribution being observed in the experiment. As we shall see, the unimode

nature of the distribution can be understood by treating more clearly the ground state spin degeneracy.

We calculate the ground state energies for a system of interacting electrons modeled by a tight-binding 2D Hamiltonian given by:

$$H = \sum_{k,j,s} \epsilon_{k,j} a_{k,j,s}^\dagger a_{k,j,s} - V \sum_{k,j,s} (a_{k,j+1,s}^\dagger a_{k,j,s} + a_{k+1,j,s}^\dagger a_{k,j,s} + h.c) + H_{int}, \quad (2)$$

where $\epsilon_{k,j}$ is the energy of a site (k, j) , chosen randomly between $-W/2$ and $W/2$ with uniform probability, V is a constant hopping matrix element, and $s = \uparrow, \downarrow$ is the spin. The interaction Hamiltonian is given by:

$$H_{int} = U' \sum_{k,j} a_{k,j,\uparrow}^\dagger a_{k,j,\uparrow} a_{k,j,\downarrow}^\dagger a_{k,j,\downarrow} + U \sum_{k,j>l,p;s,s'} \frac{a_{k,j,s}^\dagger a_{k,j,s} a_{l,p,s'}^\dagger a_{l,p,s'}}{|\vec{r}_{k,j} - \vec{r}_{l,p}|/b} \quad (3)$$

where U' is the on-site interaction constant and $U = e^2/b$ is the long range component of the Coulomb interaction constant where b is a lattice constant.

We consider a 4×3 dot with $M = 12$ sites and up to $N = 9$ electrons. The size of the many-body Hilbert space is $m = \binom{2M}{N}$, thus for $M = 12$ and $N = 9$ we end up with $m = 1,307,504$. One can use the fact that in the site representation the Hamiltonian matrix has no off-diagonal terms which couple states of different S_z (where S_z is the component of the total spin in the \hat{z} direction) to diagonalize each block with a given S_z separately. The size of each block is $\binom{M}{N_\uparrow} \times \binom{M}{N_\downarrow}$ which for the largest case is equal to 392,040. After the minimal eigenvalue for each block is obtained the global minimal eigenvalue is found. Since the system has a $2S + 1$ ground state degeneracy (where S is the total ground state spin), for all the cases in which the total ground state spin $S \neq 0$ all the blocks corresponding to $S_z = -S, -S + 1 \dots S$ have the same minimal eigenvalue. This gives an excellent check for the accuracy of the diagonalization procedure. Therefore, both the ground state energy and the value of S are found by the exact diagonalization.

The strength of the long range component of the Coulomb interactions, U , is varied between $0 - 6V$. For the results presented here, an on-site coupling $U' = (10/3)U$ was chosen, corresponding to Hubbard's calculation of the ratio of U' to U for weakly overlapping

hydrogen like wave-functions [11]. As will be discussed later the exact ratio of U' to U does not play a crucial role in determining the main results presented here. The disorder strength is set to $W = 8V$ in order to assure perfect RMT behavior for the non-interacting case. We also present some data pertaining to the $W = 3V$ case, where only the lower levels (up to $N = 6$) which show RMT behavior were considered in order to gain some insight into the role of disorder. For each value of U , the results for each value of N are averaged over 500 different realizations of disorder.

The results of the tight-binding calculations at a given interaction strength U can then be compared to the experimental quantum dot density parameters. The ratio of the average inter-particle Coulomb interaction and the Fermi energy $r_s = 1/\sqrt{\pi n}a_B$ (where n is the electronic density and a_B is the Bohr radius) corresponds to $r_s \sim \sqrt{\pi/2}(U/4V)$ for $N = 6$, $M = 12$. For all experimental setups $r_s \sim 1$ [1,3], resulting in $U \sim 4V$.

The average value $\langle\Delta_2\rangle$ is presented in Fig. 1. In the non interacting case ($U = 0$), $\langle\Delta_2\rangle = 0$ for even electron numbers, while for odd numbers $\langle\Delta_2\rangle = \Delta$. Even for weak interactions ($U = 0.6V$ corresponding to $r_s \sim 0.2$) there is no remnant of the even-odd asymmetry in $\langle\Delta_2\rangle$. Moreover, up to $U \sim 4$, for all values of N , $\langle\Delta_2\rangle = e^2/C + \Delta'$, where the capacitance $e^2/C = 0.67U$ was calculated through the random phase approximation (for details see Ref. [12]) and has no adjustable parameters, and $\Delta' = 0.81\Delta$ is obtained from a fit. Since this behavior appears already at weak interactions and the influence of the long range component of the Coulomb interaction is well described by the capacitance up to $U \sim 4V$ ($r_s = 1$) it is natural to concentrate on the role of the on-site interactions. In first order perturbation theory for the on-site interaction strength, where the single electron eigenfunctions are assumed to follow the random vector model (RVM), it has been shown [13] that $\langle\Delta_2\rangle = e^2/C + 3U'/(M+2)$ for an even number of electrons and $\langle\Delta_2\rangle = e^2/C + \Delta - 2U'/(M+2)$ for the odd case. Thus, the even and odd values of $\langle\Delta_2\rangle$ will coincide at $U'_c = (M+2)\Delta/5 \sim 8V/5$, which corresponds here to $U = 0.5V$, in good agreement with the numerical results. We have checked that this value of U'_c holds for different ratios between U and U' . Thus, even for the smallest conceivable ratio ($U' = U$, since $U' < U$

represents a locally attractive interaction) the even-odd asymmetry will disappear for $r_s > 0.4$, while a more reasonable estimation of the ratio will yield the values quoted in the introduction. Above this value of interaction it is not possible to continue to use this first order perturbation, but it is reasonable to assume that the excess value of $\langle\Delta_2\rangle$ above e^2/C will be of order $\Delta' = 3U'_c/(M+2)$, i.e., $\Delta' \sim 0.6\Delta$, which is not far from the numerical result. Above $U \sim 4V$ ($r_s = 1$) short range correlations in the electronic density appear and the random phase approximation is no longer valid, as discussed in Refs. [1,12].

The fluctuations $\langle\delta^2\Delta_2\rangle = \langle(\Delta_2)^2\rangle - \langle\Delta_2\rangle^2$ are portrayed in the inset of Fig. 1. As with the average, the even-odd asymmetry disappears at $U = 0.6V$ and a gradual enhancement of the fluctuations as a function of the interaction strength is seen. The full distribution of Δ_2 obtained for all values of N ($N = 4 \dots 9$) is shown in Fig. 2. In Fig. 2a the distribution is plotted as function of $\Delta_2 - e^2/C$, which in a sense captures the distribution of the “single electron level” spacings. In the non-interacting case the CI+RMT distribution (Eq. 1) fits perfectly. Even for weak interactions, $U = 0.6V$ ($U' = 2V$), the bimodal distribution is wiped away. Neither does the SR-RMT distribution [3,9] fit. The best fit for weak interactions is obtained from the usual RMT distribution

$$P_{RMT}(s) = (\pi s/2) \exp(-\pi s^2/4) \quad (4)$$

where $s = (\Delta_2 - e^2/C)/\langle\Delta_2 - e^2/C\rangle$. This feature does not seem to depend much on the disorder, as can be seen from the $W = 3V, U = 0.6V$ curve in Fig. 2a. As the interaction strength increases, the distribution becomes wider, less asymmetric, and from the fact that a considerable weight of the distribution is at negative values it becomes clear that no constant interaction model is able to describe it. For the region of $r_s \sim 1$ ($U \sim 4V$) a better fit of the distribution to a Gaussian is obtained. The same results plotted as function of $\Delta_2/\langle\Delta_2\rangle$ are shown in Fig. 2b. A clear crossover from the RMT like distribution to a Gaussian distribution is seen as the interaction is increased, although its width does seem to depend on the disorder (see the $W = 3V, U = 3.6V$ curve in Fig. 2b).

In all experiments a distribution which is Gaussian (up to deviations in the tail) is seen

[1–3]. Since the experiments are performed in the region of $r_s \sim 1$ this is in good agreement with our results. Leaving aside the width of the distribution for the moment, we need to understand better the unexpected disappearance of the bimodal distribution for intermediate coupling. We can gain insight into this by studying the ground state polarization of the dot. In the non-interacting case we expect the total spin of the dot to be $S = 0$ for an even number of electrons and $S = 1/2$ for an odd number. For weak interactions, using first order perturbation in the on-site interaction strength with the RVM one finds that on the average the system gains $3U'/(M + 2)$ interaction energy by flipping one spin, while it loses $\eta_{N/2+1} - \eta_{N/2}$ kinetic energy for an even number of electrons, compared to the gain of $4U'/(M + 2)$ interaction energy and the loss of $\eta_{(N+3)/2} - \eta_{(N-1)/2}$ kinetic energy for an odd number. The dot thus will flip a spin if the gain in interaction energy will be larger than the loss of kinetic energy [13]. The probability of finding two consecutive small single electron level spacings is much smaller than the probability of finding one small level spacing, therefore we expect a finite probability for an even number of electrons to be in a $S = 1$ state, while a much lower probability to find an odd number of electrons at $S = 3/2$ is expected. Indeed in Fig. 3a it can be seen that while the average spin of the ground state for an odd number of electrons is $\langle S \rangle \sim 1/2$, except for strong interactions $U > 4V$ ($r_s > 1$), the average spin for the even case $\langle S \rangle > 0$ also for rather weak interactions. Since both the interaction energy and the kinetic energy scale as $1/M$ we expect this behavior to hold also for large systems. Thus, in the $r_s < 1$ region we expect to see a significant number of $S = 1$ states for an even number of electrons and almost no $S = 3/2$ states for an odd number. Higher spin polarizations seem to be rather rare, although we encountered two realizations with $S = 2$ for $N = 8$ and $U = 1.8V$. Once interactions are stronger the even odd asymmetry in the ground state spin polarizability is less pronounced. This is illustrated in Fig. 3b, where $\langle |S_N - S_{N-1}| \rangle$ is plotted. For any non-correlated behavior $|S_N - S_{N-1}| = 1/2$, while for correlated behavior adding an electron to the dot may flip the spin of other electrons already in the dot and even lead to spin blockade [14]. A clear indication for the appearance of a correlated state is seen for $r_s > 1$.

Recently, Stopa [15] has suggested that spin polarization in a chaotic dot may appear due to scar states. According to this scenario the scar state will be populated first by an \uparrow electron, then several other states of higher kinetic energy will be populated in the regular sequence and eventually a \downarrow will populate the scar state. This scenario leads to a shift by a $1/2$ of S_N between populating the scar state with the \uparrow electron and the \downarrow electron. This is a distinct spin polarization pattern from the ones previously described. The combined effect of scar states and regular chaotic or disordered states considered in this letter is an interesting question now under investigation. A very recent paper [16] has suggested that the absence of the bimodal distribution is due to the deformation of the confining potential of the dot as electrons are added [17]. Again such a scenario will lead to a different spin polarization pattern than the previous ones, i.e., it will be equivalent to the non-interacting one. The measurement of the dot's spin polarization is feasible [18], probably via measuring the magnetic field dependence of the differential conductance [19], from which the change in dot's spin may be observed. As shown in this letter, by studying the change in the spin polarization it is possible to clarify whether the underlying physics of the dot corresponds to the correlated regime, the intermediate regime, a deformable potential, or maybe carries the signature of scar states.

There remains a question regarding the width of the distribution. While earlier experiments show a width comparable to $0.15 - 0.10e^2/C$ [1,2], a recent experiment [3] yields a width of $0.05e^2/C \sim \Delta$. Our simulations show $\sqrt{\langle \delta^2 \Delta_2 \rangle} \sim 0.2e^2/C$ for $W = 8V$ and $\sqrt{\langle \delta^2 \Delta_2 \rangle} \sim 0.1e^2/C$ for $W = 3V$, consistent with previous results for spinless electron of $\sqrt{\langle \delta^2 \Delta_2 \rangle} \sim 0.1 - 0.2e^2/C$, and with the earlier experiments [1,2]. Nevertheless, for the numerical study at $U = 4V$ ($r_s = 1$), $0.2e^2/C \sim \Delta$, which does not contradict Ref. [3]. It is clear that for $r_s \ll 1$, $\sqrt{\langle \delta^2 \Delta_2 \rangle} \propto \Delta$, while for $r_s \gg 1$, $\sqrt{\langle \delta^2 \Delta_2 \rangle} \propto e^2/C$. Whether the behavior of the fluctuations in the above model at $r_s \sim 1$ is determined by Δ or e^2/C , and the role of disorder, could be determined only by a careful finite size study, since the change in size will change the ration of Δ to e^2/C and enable the determination of the relevant scale.

In conclusion, the on-site interaction is responsible of the removal of the even-odd asymmetry in the addition spectrum distribution. These interactions lead to a ground state spin polarizability of the dot. The dependence of the spin polarizability on the number of electrons may be used as a sensitive tool to determine the relevant physics in the dot.

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FIGURES

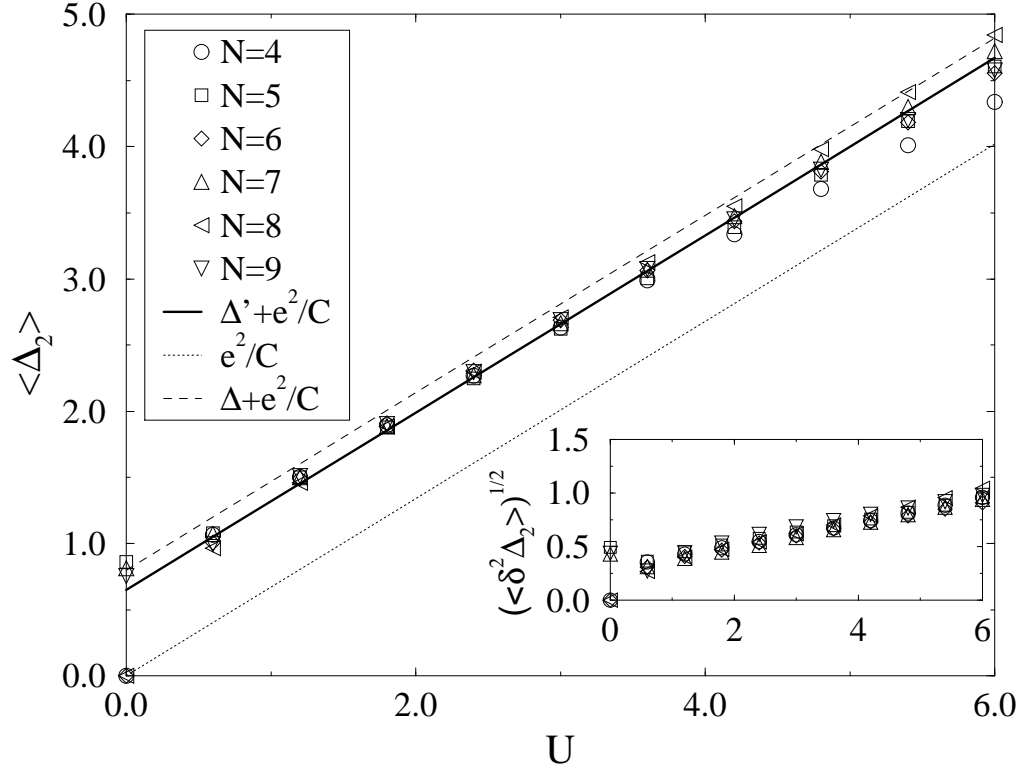


FIG. 1. The value of $\langle \Delta_2 \rangle$ as function of U for a 4×3 lattice and a different number of electrons N . All values are in units of the hopping matrix element V . Inset: the fluctuations .

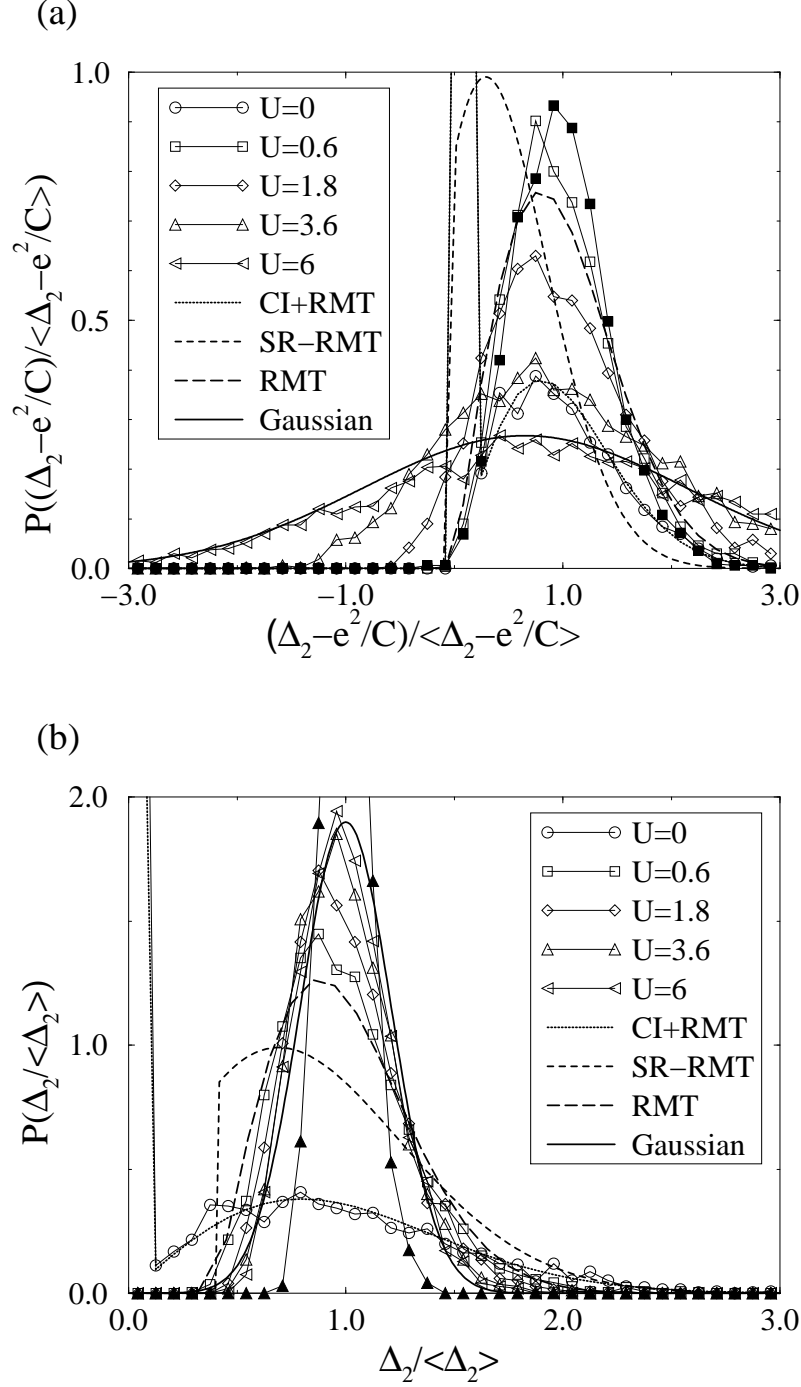


FIG. 2. The probability distribution of Δ_2 for different values of the interaction U , drawn on the scale of (a) $\Delta_2 - e^2/C$ and of (b) Δ_2 . The heavy lines correspond to Eq. (1) with $e^2/C = 0$, SR-RMT, Eq. (4) and to a Gaussian with a variance of $0.2e^2/C$. Open symbols correspond to $W = 8V$ while filled symbols to $W = 3V$.

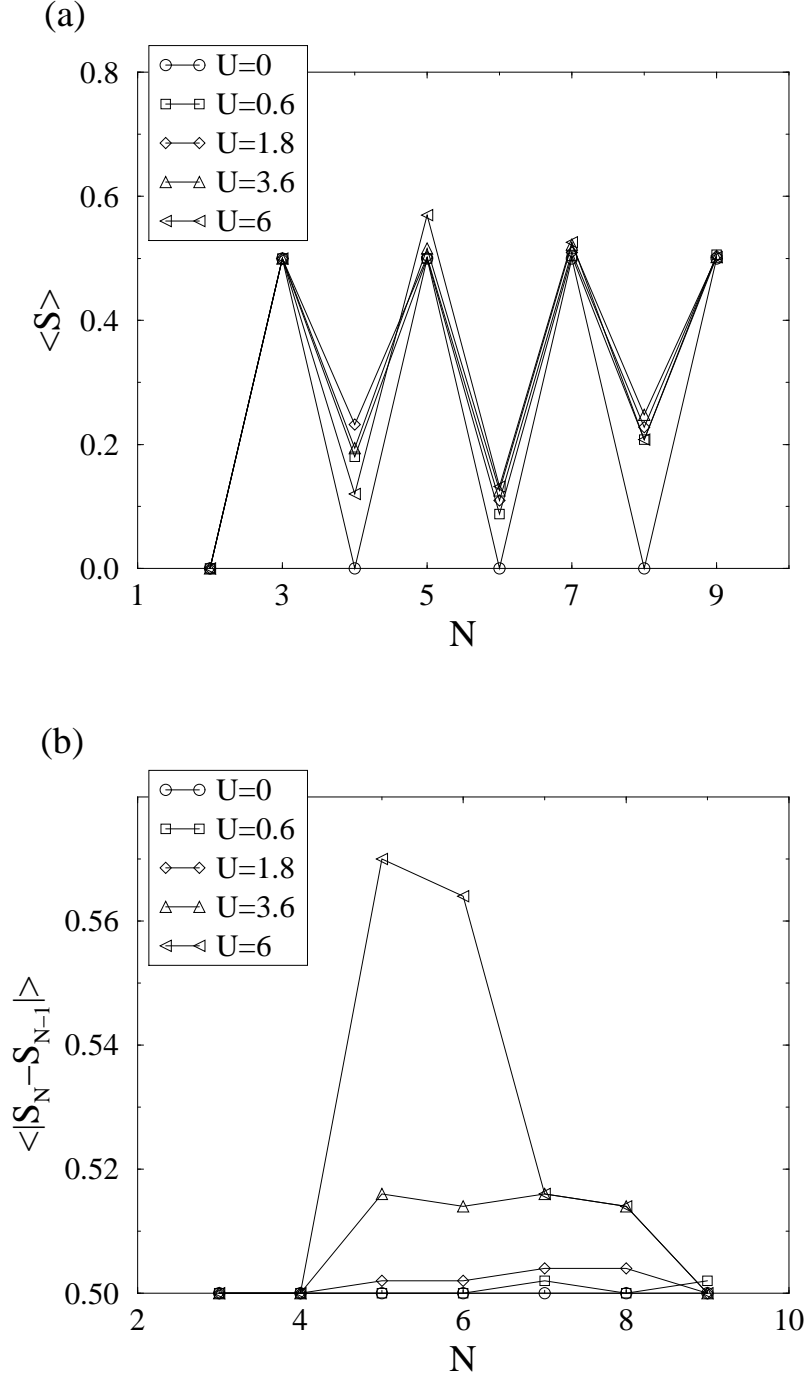


FIG. 3. (a) The ground state average spin, and (b) the average of the absolute value of the change in the ground state spin as an electron is added to the dot, as function of the number electrons for different interaction strength.